IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE HONORABLE BOARD OF PATENT APPEALS AND INTERFERENCES

Appl. No. : 10/576,299 Confirmation No. 4456

Applicant : Motoshige SUMINO et al.

Filed : April 19, 2006

TC/A.U. : 1621

Examiner : Chukwuma O. Nwaonicha

Dkt. No. : WKP-003

Cust. No. : 20374

I hereby certify that this paper is being facsimile transmitted on April 21, 2009, to the United States Patent and Trademark Office to facsimile number (571)273-8300.

Ronald J. Kubovcik

BRIEF ON APPEAL

Mail Stop Appeal Brief - Patents
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

April 21, 2009

Sir:

This is an appeal from the decision dated July 21, 2008, of the primary Examiner finally rejecting claims 1-8 and 10 in this application.

(i) REAL PARTY IN INTEREST

The real party in interest is Wako Pure Chemical Industries, Ltd., Osaka, Japan.

(ii) RELATED APPEALS AND INTERFERENCES

There are no related appeals or interferences which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

(iii) STATUS OF CLAIMS

Claims 1-8 and 10 are pending in this application. Claim 9 has been cancelled. Claims 1-8 and 10 are appealed. Claims 1-8 and 10 as finally rejected appear in the attached Claims Appendix.

(iv) STATUS OF AMENDMENTS

No amendments were filed subsequent to the final rejection.

(v) SUMMARY OF CLAIMED SUBJECT MATTER

The subject matter defined in claim 1, the only independent claim involved in the appeal, is a method for producing a triarylsulfonium salt having a structure in which only one of the three aromatic rings on the cation portion of the salt is different from the other two aromatic rings (specification, page 1, lines 6-10; and page 3, lines 18-24).

In the method of the present invention for producing the triarylsulfonium salt, a diaryl sulfoxide and an aryl Grignard

reagent are reacted (Specification, page 4, lines 10-15) in the presence of an activator with high affinity for oxygen (hereinafter, referred to simply as the activator) in an amount of 4.5 to 7.5 equivalents relative to the the diaryl sulfoxide. (The upper limit of 7.5 equivalents is supported, inter alia, in the specification on page 4, lines 17-18, and the lower limit of 4.5 equivalents is supported, inter alia, on page 38, line 10-14; and page 47, lines 5-8). The resultant reaction mixture is then reacted with a strong acid, or a salt thereof (specification, page 4, lines 18-20).

The method of the present invention shows unexpected excellent effects with the use a larger amount of the activator than has been conventionally used. In particular, the use of 4.5 to 7.5 eq. of the activator can obtain the desired sulfonium salt at a high yield and without byproducts (specification, page 5, lines 3-11; page 46, Table 5, Experimental Examples 3 to 6).

The method of the present invention has been made to solve the problem that when a diaryl sulfoxide [two aromatic rings have the same structure (structure [a])] and an aryl Grignard reagent having an aromatic ring different from the aromatic ring of the diarylsulfoxide (structure [b]) are reacted in the presence of an activator, not only the desired compound (wherein two aromatic

rings are structure [a] and one aromatic ring is structure [b]) is obtained, but also two kinds of byproduct are obtained. One byproduct is a sulfonium salt wherein three aromatic rings are all structure [a], and the other byproduct is a sulfonium salt wherein one aromatic ring is structure [a] and two aromatic rings are structure [b]). (specification, page 2, lines 12-24).

The fact that the desired sulfonium salt can be efficiently produced in a high purity without byproducts is extremely important.

First, when the triarylsulfonium salt not containing byproducts obtained by the method of the present invention is used as a photo acid-generating agent in a photolithography step in the manufacture of a semiconductor, an improvement of roughness on a profile or a sidewall of a hyperfine pattern and formation of a good rectangle pattern of reduced edge roughness is obtained (specification, page 40, lines 13).

In contrast, when by-products are contained in a product, use of such a product as an acid generator changes the efficiency of the acid generation. In other words, sensitivity (e.g., acid generation efficiency) to exposure is different between the product (objective compound) and by-products because the structure of the product and by-products is different. And when a product

containing by-products, in which by-product content undergoes a change with each production lot, is used as an acid generator, it is difficult to quantify the amount of acid generation.

As a result, formation of a resist pattern with the use of such an acid generator causes problems such as poor-reproducibility of the pattern.

Formation of a hyperfine pattern is leading-edge technology. Therefore, for formation of a resist pattern with good reproducibility, use of a product with high purity as an acid generator is required.

Second, separation of by-products from the desired triarylsulfonium salt is difficult because the structure of the by-products is similar to that of the desired product.

The separation of the by-product from the desired product, as would be understood by one of ordinary skill in the art, requires the use of a preparative liquid chromatography. However, a preparative liquid chromatography is a separation and refinement means normally used on a laboratory scale. It is difficult to use preparative liquid chromatography as a separation means on an industrial-scale due to a limited amount of one preparation isolation.

(vi) GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

(A) Whether claims 1-8 and 10 are unpatentable under 35 U.S.C. 103(a) over Oono et al., U.S. Patent No. 6,723,483 ("Oono"), in view of Osawa et al., U.S. Patent No. 5,824,824 ("Osawa").

(vii) ARGUMENT

Rejection under 35 U.S.C. § 103(a) over Oono in view of Osawa

The Claimed Range of Activator of 4.5 to 7.5 Equivalents Relative to the Diaryl Sulfoxide Is Critical to Obtaining the Desired Triarylsulfonium Salt Without Byproducts

The present invention, as defined in claim 1, is a method for producing a triarylsulfonium salt having a structure in which only one of the three aromatic rings on the cation portion of the salt is different from the other two aromatic rings. In the method of the invention a diaryl sulfoxide, in which the two aromatic rings have the same structure (structure [a]), and an aryl Grignard reagent in which the aromatic ring (structure [b]) is different from the aromatic ring of the diaryl sulfoxide, are reacted in the presence of an activator.

In a method such as in the present invention where a diaryl sulfoxide, in which the two aromatic rings have the same structure (structure [a]), and an aryl Grignard reagent having an aromatic ring (structure [b]) different from the aromatic ring of the

diarylsulfoxide, are reacted in the presence of an activator, in addition to the desired compound (wherein two aromatic rings are structure [a] and one aromatic ring is structure [b]), two kinds of byproduct can be obtained. One byproduct is a sulfonium salt wherein three aromatic rings are all structure [a], and the other byproduct is a sulfonium salt wherein one aromatic ring is structure [a] and two aromatic rings are structure [b]). Such byproducts are not desirable.

Appellants have discovered that the formation of the byproducts can be prevented by using an amount of activator of 4.5 to 7.5 equivalents relative to the diaryl sulfoxide. Neither Oono nor Osawa nor the combination thereof discloses or suggests that the amount of activator is critical to eliminating byproducts.

The criticality of the amount of activator is demonstrated by the comparative data in the present application. In particular, Table 5 on page 46 of the specification describes the yields of the obtained objective compound and those of the byproducts obtained when using chlorotrimethylsilane (TMSCl) as the activator in amounts of 2.5, 3.0, 4.0, 5.0, 6.0, 7.0, 7.5 eq., relative to the diphenyl sulfoxide.

As is demonstrated by the data of Table 5, the use of TMSCl as the activator in an amount of 2.5 eq. relative to the diphenyl

sulfoxide forms not only byproducts but also a low yield of the objective compound (59%) (see Comparative example 1). The use of TMSCl (activator) in an amount of 3 and 4 eq. as in Experimental Examples 1 and 2 produces the objective compound with a high yield (72%), but still results in the formation of byproduct.

In contrast, as shown by the results of Experimental Examples 3 to 6, use of TMSCl (activator) in an amount of 5, 6 and 7.5 eq. relative to the diphenyl sulfoxide does not result in the formation of byproducts at all and still produces the objective compound with a high yield.

The Results of the Use of an Amount of Activator of 4.5 to 7.5 Equivalents Relative to the Diaryl Sulfoxide Are Unexpected Relative to the Prior Art

Oono

Oono discloses a method for producing a triarylsulfonium salt by reacting diaryl sulfoxide, trimethylsilylsulfonate (an activator) and an aryl Grignard reagent (see Oono, column 11, lines 45-48).

However, the method of Oono for producing a sulfonium salt, different from the method of the present invention, includes the production of a salt whose cation portion has three identical aromatic rings.

An amount of activator to be used in Oono is 0.8 to 2 mol

relative to 1 mole of the diarylsulfoxide (0.8 to 2 eq. relative to the diaryl sulfoxide).

In the examples of Oono, there is a disclosure of a method for producing a sulfonium salt in which the cation portion has two identical aromatic rings (structure [a]) and one aromatic ring (structure [b]) different from the other two rings using a trimethylsilyl sulfonate, such as trimethylsilyl triflate, in an amount of 0.13 mol relative to diphenyl sulfoxide of 0.1 mol (i.e., an amount of the activator of 1.3 eq. relative to the diphenyl sulfoxide). The yield of the sulfonium salt is low (43%). There is no description of the amount of byproduct(s). However, the results of the experimental examples in the present specification as discussed above indicate that byproducts are formed in the example of Oono.

Osawa

Osawa discloses a method for producing a triarylsulfonium salt by reacting a diaryl sulfoxide and an aryl Grignard reagent in the presence of trimethylsilyl chloride (activator) of 1 to 5 mole, preferably 2 to 3 mole, relative to the diaryl sulfoxide (see column 11, line 32, to column 12, line 4).

Similar to Oono and different from the present invention, the sulfonium salts obtainable by the method of Osawa includes a

sulfonium salt having three identical aromatic rings on the cation portion.

In Osawa there are examples of the synthesis of a sulfonium salt having two identical aromatic rings (structure [a]) and one aromatic ring different from the other two (structure [b]). (See Osawa, synthesis examples 2 and 4). However, there is no description relating to the formation of byproducts. I.e., there is neither a disclosure nor a suggestion of steps to be taken in order to avoid the formation of both a byproduct having three identical aromatic rings (structure [a]) and a byproduct having one aromatic ring (structure [a]) and two aromatic rings (structure [b]) of the three aromatic rings.

Osawa discloses that the activator trimethylsilyl chloride (TMSCl) can be used in an amount of 1 to 5 eq., but describes that an amount of 2 to 3 eq. relative to the diaryl sulfoxide is especially preferred. In the examples of Osawa an amount of TMSCl of 2.5 or 3 eq., relative to the diaryl sulfoxide, is used. (See Osawa, column 17, synthetic example 1; column 18, synthetic example 2; and column 20, synthetic example 4). There is no description of amounts and/or kinds of byproduct produced in the examples. However, as shown by the results of the experimental examples in the present specification, the use of TMSCl in a range of 2.5 to 3

eq. as used in the examples of Osawa results in the production of byproducts.

Combination of Oono and Osawa

The combination of Oono and Osawa fails to disclose or suggest that in the production of a triarylsulfonium salt having a structure in which only one of the three aromatic rings on the cation portion of the salt is different from the other two aromatic rings, the production of undesired sulfonium salts can be avoided by using an amount of activator of 4.5 to 7.5 equivalents relative to the diaryl sulfoxide.

The criticality of the characteristic feature of the present invention and the excellent effects obtained by this feature, that is, the use of an activator such as TMSCl of 4.5 to 7.5 eq. relative to the diaryl sulfoxide to obtain the desired compound in yields comparable to the prior art, but **without the production of byproducts**, cannot be reasonably predicted based on the combination of Oono and Osawa and supports the non-obviousness of the method of the present invention.

Advisory Action

In the Advisory Action dated February 3, 2009, the Examiner acknowledges appellants argument that the use of 4.5 to 7.5 eq. of the activator can obtain a desired sulfonium salt efficiently in a

high purity without byproducts (Advisory Action, page 2, lines 3-4). However, the Examiner then concludes:

> "Applicants' arguments have been consider [sic] in light of the prior art references cited and the Examiner notes that there is no significant improvement in terms of unexpected result provided by the current process. The difference in terms of product yield between the current application and the process of the prior art references cited is insignificant to warrant unexpected result [sic]."

(Advisory Action, page 2, lines 4-7) (Emphasis added).

The yield of the desired triarylsulfonium salt in the method of the present invention is not the unexpected result. The unexpected result is the production of the desired triarylsulfonium salt (in yields which, as acknowledged by the Examiner in the Advisory Action, do not differ significantly from the prior art references) without byproducts.

Reversal of the rejection of claims 1-8 and 10 as being unpatentable under 35 U.S.C. 103(a) over Oono et al., U.S. Patent No. 6,723,483, in view of Osawa et al., U.S. Patent No. 5,824,824, is in order and is respectfully requested.

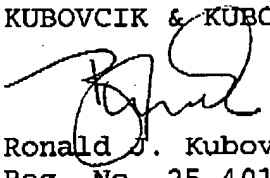
In view of the foregoing argument, appellants respectfully

requests that the Final Rejection of the Primary Examiner be reviewed and reversed.

Please charge any required fees or credit any overpayment to our Deposit Account No. 111833.

Respectfully submitted,

KUBOVCIK & KUBOVCIK



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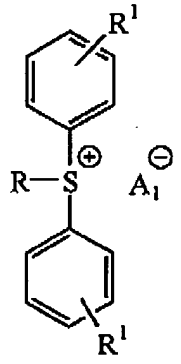
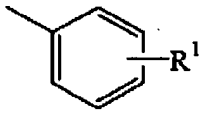
(viii) CLAIMS APPENDIX

1. A method for producing a triarylsulfonium salt represented by the general formula [4]:



[4]

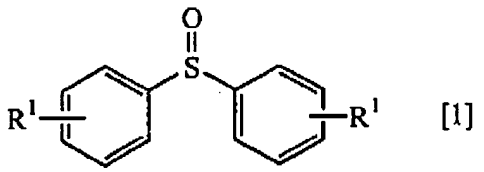
wherein, the two R^1's are the same and are hydrogen atom, halogen atom, alkyl group, haloalkyl group having 1 to 4 carbon atoms, alkoxy group, acyl group, hydroxyl group, amino group, nitro group or cyano group; R represents an aryl group which may have a substituent selected from a halogen atom, an alkyl group, a haloalkyl group having 1 to 4 carbon atoms, an alkoxy group, an alkylthio group, a N-alkylcarbamoyl group and a carbamoyl group, with the proviso that R is different from



;

and A_1 represents a strong acid residue,

comprising reacting a diaryl sulfoxide represented by the general formula [1]:



[1]

and an aryl Grignard reagent represented by the general formula [2]:

RMgX [2]

wherein, X represents a halogen atom; R represents the same as above,

in the presence of an activator with high affinity for oxygen of 4.5 to 7.5 equivalents relative to the above diaryl sulfoxide, and then reacting the resultant reaction mixture with a strong acid represented by the general formula [3]:

HA_1 [3]

wherein, A_1 represents the same as above,

or a salt thereof.

2. The method according to claim 1, wherein the activator with high affinity for oxygen is a halogenotriorganosilane.

3. The method according to claim 1, wherein the activator with high affinity for oxygen is a halogenotrialkylsilane.

4. The method according to claim 1, wherein the activator with high affinity for oxygen is chlorotrimethylsilane.

5. The method according to claim 1, wherein the amount of use of an activator with high affinity for oxygen is 1.2 to 3 equivalents relative to the aryl Grignard reagent represented by the general formula [2].

6. The method according to claim 1, wherein a strong acid residue represented by A_1 is an anion derived from a hydrohalic acid represented by the general formula [5]:

HX_1 [5]

wherein, X_1 represents a halogen atom,

a sulfonic acid represented by the general formula [6]:

R^2-SO_3H [6]

wherein, R^2 represents an alkyl group, an aryl group or an aralkyl group, which may have a halogen atom, or a camphor group,

or an inorganic strong acid represented by the general formula [7]:

HM_1Fn [7]

wherein, M_1 represents a metalloid atom; and n represents 4 or 6.

7. The method according to claim 6, wherein X_1 is a chlorine atom or a bromine atom.

8. The method according to claim 6, wherein the metalloid atom represented by M_1 is a boron atom, a phosphorus atom, an arsenic atom or an antimony atom.

10. The method according to claim 1, wherein the reaction of the diaryl sulfoxide and the aryl Grignard reagent is conducted in the presence of the activator of 5 to 7.5 equivalents relative the diaryl sulfoxide.

(ix) EVIDENCE APPENDIX

None

(x) RELATED PROCEEDINGS APPENDIX

None